

**Method of manufacturing a diamond composite body having a modified outer surface.**

5      **TECHNICAL FIELD**

The present invention relates to a method of manufacturing a diamond composite body comprising the steps of forming a porous body containing diamond particles and graphite and/or carbon and infiltrating silicon or silicon alloy into the body and a body produced by the method.

**BACKGROUND OF THE INVENTION**

Diamond composites have several excellent properties like high hardness; wear resistance and stiffness. Special variants like materials manufactured in accordance with WO 02/42240 also has high thermal conductivity, typically 600 W/mK. These properties make some of these materials suitable for different applications like inserts for machining of materials like metals, rock, concrete, wood etc. For material grades with high thermal conductivity diamond composites can be excellent as heat sinks and heat spreaders in electronic applications.

A drawback of the application of these materials has been the joining methods. When using conventional methods of brazing, soldering, clamping and gluing the advantages of diamond composites are partly lost. This is due to several reasons. One reason is the bad wetting behaviour of the surface of the composites to most other interface materials. A second reason is the comparatively rough surface obtained. A third reason is the difficulty to obtain very flat surfaces.

Thermal conductivity is often lost in the joining material and in the joining surfaces. The standard processes of producing diamond composites do not create very flat surfaces, which are desirable for limiting the thickness of the comparatively low conducting joining layer. The machining of diamond composites is extremely difficult and expensive. Many diamond composites are also very difficult and expensive to machine to a smooth surface, at a reasonable cost. The roughness of the surface can make it more difficult to make a good joint.

A problem with diamond composite materials is thus that it is difficult to join these materials to tools and electronic appliances with conventional methods of brazing, soldering, clamping and gluing.

The objective of the invention is to solve this problem.

## SUMMARY OF THE INVENTION

This objective is solved by a method of manufacturing a diamond composite body comprising the steps of forming a porous body containing diamond particles and graphite and/or carbon and infiltrating silicon or silicon alloy into the body and heating the body to form carbide by reaction between graphite or carbon and the infiltrated silicon or silicon alloy, characterised by the further steps of providing a surplus of silicon or silicon alloy in connection with the infiltration step so that a layer of silicon or silicon alloy will cover at least one outer surface of the composite body. By this method a body having a surface that can more easily be joined to other components with conventional methods is obtained.

In a preferred embodiment the at least one outer surface covered with silicon or silicon alloy is thereafter machined to be flat.

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In a preferred embodiment for bodies with flat surface, the at least one outer surface is covered with silicon or silicon alloy that is preferably machined to a flatness less than 0.1 mm and a surface roughness of less than Ra 0.01 mm, typically being less than Ra 0.002 mm. Advantageously, the surplus of silicon or silicon alloy is supplied in such an amount that the layer of silicon or silicon alloy covering the at least one outer surface of the composite body with a thickness of 0.01-2 mm, preferably 0.05-0.1 mm, before machining. The layer of silicon or silicone alloy on the at least one outer surface is preferably machined to a thickness of 0-1 mm.

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In another embodiment a silicon alloy comprising at least one element from the group consisting of metals Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, Mn, Re, Co, Ni, Cu, Ag, Al and the elements B and Ge is infiltrated.

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The invention also relates to a composite body containing diamond particles bonded to a matrix of silicon or silicon alloy carbide and silicon or a silicon alloy, characterised in that at least one outer surface of the body is covered by an outer layer of silicon or silicon alloy.

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In a preferred embodiment the at least one outer surface has a flatness of less than 0.1 mm, preferably less than 0.002 mm, and a surface roughness less than Ra 0.01 mm. Advantageously, the outer layer on said at least one outer surface has a thickness of 0-1 mm.

The invention also relates to a component comprising a composite body as defined above.

## BRIEF DESCRIPTION OF THE DRAWING

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The invention will now be described with reference to the Figure 1 showing a schematic sectional view of a surface part of a material according to a preferred embodiment of the invention.

## 10 DESCRIPTION OF EMBODIMENTS

The material according to the present invention is preferably produced according to methods presented in WO99/12866 and WO00/18702. These methods include the following steps:

- 15       1. Forming a porous workpiece out of a mixture containing diamond particles.
2. Heat treating the work piece and controlling the heating temperature and heating time so that a certain desired amount of graphite is created by graphitization of diamond particles.
- 20       3. Infiltrating melted silicon or alternatively a silicon alloy into the work piece.
4. Reacting of the molten silicon or silicon alloy and graphite to form SiC or SiXC.

25       By the manufacturing process described above an article with a predetermined shape is formed.

The work piece is formed out of a mixture of diamond particles preferably consisting of at least two different fractions with different diamond particle sizes. Of the diamond content in the workpiece at least 50 weight% should have a diameter of 80  $\mu\text{m}$  or above. The use of at least two different fractions with different diamond particle sizes is advantageous in order to reach a packing degree in the work piece that in the sintered compact gives a high enough diamond concentration (i.e. a short path for the phonons to travel between the diamonds) to reach the required levels of thermal diffusivity and thermal conductivity.

Forming of the workpiece is carried out by conventional ceramic forming methods such as pressing, injection molding, tape casting, slurry casting or gel casting using conventional equipment.

The duration of the heat treatment of the workpiece is as is needed for the diamond particles to have been transformed to graphite to the desired amount. An example of heat treatment is heating the workpiece to temperatures between 1000 and 1900°C in vacuum or an inert atmosphere.

The infiltration of molten Si or silicon alloy is carried out by such known methods as melting a solid piece on or near the surface of the workpiece, feeding already molten Si or silicon alloy on to the surface of the workpiece or by dipping the workpiece into a melt of Si or silicon alloy. As the melt infiltrates the workpiece it reacts with graphite or carbon and forms SiC or a SiC phase including elements from the alloying elements. The formed silicon carbide phase and a small amount of un-reacted silicon or silicon alloy phase fill up the porous space of the workpiece.

The heat treatment and infiltration can be carried out in separate heating steps or in a continuous heating cycle.

The infiltrating melt used can be a silicon alloy comprising at least one metal from the group consisting of Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W, Mn, Re, Fe, Co, Ni, Cu, Ag, Al, and the elements B or Ge. In this case small amounts of secondary phase compounds may form, such as metal silicides, metal carbides, etc.

The sintered composite material thus consists of three major phases, a diamond phase, a carbide phase around the diamond particles and un-reacted silicon or silicon alloy phase between areas of carbide. The carbide that has formed from the reaction between the graphitized diamond and the melt is coating and surrounding each individual diamond particle. The carbide phase forms an interconnected skeleton structure, which is enclosing the diamond particles. The un-reacted silicon or silicon alloy together with the possible small amounts of secondary phase compounds, such as metal carbides, metal silicides, etc, are mainly located in the areas in-between the silicon carbide that enclose the diamond particles.

The graphite layer formed on the surface of the diamond particles has a very good adhesion to the diamond since the graphite is transformed diamond. When silicon or silicon alloy melt reacts with the said graphite the carbide formed inherits the very good adhesion to the diamond and a strong bond between the carbide and diamond is formed. When nucleation of carbide takes place on a graphite surface that has formed through graphitization of diamond the formed carbide grows epitaxially, i.e. the growth of carbide on the diamond follows the crystallographic orientation of the diamond. The manner in which carbide is formed and the strong bond between the diamond particles

and the surrounding carbide are believed to be decisive factors for obtaining the surprisingly high thermal conductivity of a material according to the preferred embodiment of the present invention. A long free path for the phonon transport is obtained in the material.

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The graphitization of the diamond surfaces has a positive effect with regards to physical surface defects on the diamond particles. The graphitization, transforms defective layers on the diamond surface to, resulting in improvement of the phonon transport path.

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Surprisingly we have found that a direct bonding between diamonds is not needed to achieve good thermal conductivity. To have a phonon transport path of high quality is more essential.

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A drawback with diamond composite materials is that they are hard to attach to other materials with conventional joining methods. When using brazing, soldering, clamping or gluing the advantages of diamond composites are partly lost due to the poor wetting properties of surface of the composite to most other interference materials. The stiffness of a construction, especially at elevated temperature, decreases for many of the joining methods. Thermal conductivity is often lost in the joining material. Moreover, the methods of manufacturing such materials do not create very flat surfaces, which is desirable for obtaining good joints and maintained thermal conductivity. However, the machining of diamond composites is extremely difficult and expensive. Many diamond composites are almost impossible to machine to a flat and smooth surface, at a reasonable cost.

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In order to remedy this drawback and create a wettable and flat surface of a diamond composite, a surplus of silicon or silicon alloy is applied in the

infiltration step. By surplus is meant an amount of silicon or silicon alloy that exceeds the amount needed for the reaction of the molten silicon or silicon alloy with all graphite and possible carbon in the work piece to form carbide and for filling the pores of the final body. Such a surplus of silicon or silicon alloy will, depending on circumstances, form an outer layer of pure silicon or silicon alloy on the composite body. Such influencing circumstances are for example supporting and surrounding parts, surface tension and gravity that can be used to control the thickness of the layer and which outer surfaces should be covered. The surplus silicon or silicon alloy will form an outer layer on at least one side, typically the upper side, of the body.

It is also possible to place the porous body in a container, like a crucible, and provide surplus silicon or silicon alloy in an amount enough to completely fill up the free space in the container. In such a case a layer of silicon or silicon alloy can be provided on all outer surfaces of the composite body.

After the forming of an outer surface layer of silicon or silicon alloy in the way described, this layer is machined to be typically flat and smooth. The flatness should be less than 0.1 mm. The roughness of the machined surface layer should be less than Ra 0.01 mm. The machining can preferably be made by diamond grinding, lapping, honing or polishing. Naturally, machining can also be made to improve surface conditions of objects with non-plane surfaces.

In figure 1 an example of a diamond composite body having an outer surface layer of pure silicon is schematically shown. The surface layer of silicon is accomplished by providing a surplus of silicon in the way described above. Diamond particles 1 are embedded in a silicon carbide skeleton 2 and pure silicon is given the reference number 3. As can be seen by this figure a layer of pure silicon covers the outer surface of the body. The typical presence of



pure silicon, if a surplus of silicon had not been provided, is indicated by interrupted lines.

5 The silicon layer 3 is strongly adhered to carbide skeleton 2 and of course also to the silicon filling the pores of the body. Moreover, the silicon layer provides an excellent surface for joining methods like brazing and soldering. By the provision of such a surface, diamond composites can easily be joined to other materials.

10 An outer layer of silicon will of course decrease the thermal conductivity of the diamond composite compared to a similar composite without such a layer. It has however surprisingly been found that for a layer of silicon being less than 300 micrometer, the decrease could hardly be measured whereas the decrease will be between 10-15% for a layer of 500 micrometer.

15 Since the porosity of the porous body into which silicon or silicon alloy infiltrated is easy to determine, it is also relatively easy to determine the amount of silicon or silicon alloy needed to infiltrate all the pores of the body and also the surplus needed to create a layer of pure silicon or silicon alloy having a certain mean thickness, on an outer surface of this body. Preferably,  
20 the amount of silicon or silicon alloy used is calculated to give such a surface layer of silicon or silicon alloy a mean thickness normally not exceeding ca. 2 mm.

25 The infiltration step will be performed in vacuum or inert atmosphere. However, the atmosphere could also contain reactive atoms or molecules, such as C or N<sub>2</sub> in order to produce other beneficial outer layers than layers of pure silicon or silicon alloy. For example, Si<sub>3</sub>N<sub>4</sub> is a very good ceramic with high oxidation resistance, wear resistance, hardness, biocompatibility and chemical

stability. If a diamond composite should work under acid, base or strong oxidation environment, such a surface layer is useful. Another example is to have the N rich atmosphere influence the electrical properties of the Si layer.

5 The main advantage of the present invention is the continuous surface layer of silicon or silicon alloy that is formed on the diamond composite. This layer is a continuous continuation of the silicon or silicon alloy filling the pores from the inside of the composite to the surface.

10 Such a surface layer can be machined with conventional methods like grinding and polishing to obtain a flat and smooth surface, if necessary even mirror surface. A machined flat surface can considerably improve the thermal conductivity through a joining by better contact to another material. A well-machined surface can also increase the mechanical properties, like strength, of  
15 a joining.

Such a surface layer avoids the need for metallization of the surface of the diamond composites, as it creates an excellent surface for joining methods like brazing and soldering. For these methods the silicon surface is a better surface  
20 chemically than the original SiC/diamond/Si composite surface.

Compared to conventional surface coating methods, like plating; CVD and PVD the proposed method gives much better properties in terms of strength and thermal conductivity. One reason is the continuous layer that connects to  
25 the internal structure of the diamond composite. The method is also economically advantageous because it does not involve any extra process step, besides the final machining.

The surface layer can also alter the electrical properties of the diamond composite, like the electrical resistivity. This is often important in electronic applications.

5 By use of a silicon or silicon alloy layer on the surface of the diamond composites, one can easily control the flatness, roughness and thickness according to different applications. One of the benefits is that silicon provides a suitable surface for brazing and soldering. In other word, silicon modifies the wetting property of the diamond composite surface.

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Surface layers made by CVD and/or PVD has several limitations, such as high cost for production, relatively low adhesive strength especially with thick layers, which are also difficult to create. In the present materials the thickness and roughness of the silicon layer can be easily be controlled according to the demands of different applications.

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In addition to the methods described in the preferred embodiments, the method of providing a surplus of silicon or silicon alloy according to the present invention can of course be used for all methods of producing diamond composites using an Si or Si-alloy infiltration step.

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